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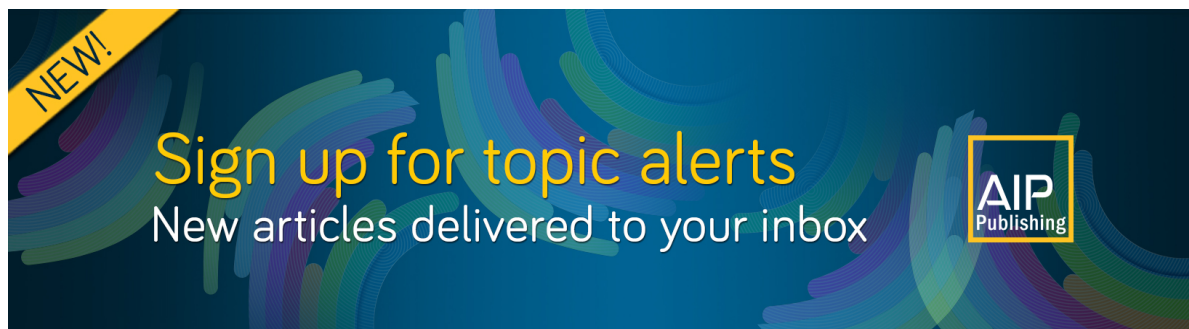
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# Increased energy of THz waves from a cluster plasma by optimizing laser pulse duration

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## ABSTRACT

We have investigated the generation of terahertz (THz) waves from an argon cluster plasma produced by laser pulses of various durations. THz energy depends on laser pulse duration and reaches a peak at a pulse duration of ~250 fs. This dependence of THz energy on laser pulse duration is attributed to the plasma density produced by the rising edge of the pulse. By irradiating clusters with collinear double laser pulses, we demonstrate that the THz energy can be increased by controlling the plasma density. Optimizing the delay time of the collinear double laser pulses increases the THz energy by ~2.5 times that with simultaneous irradiation of collinear double laser pulses.

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The generation of terahertz (THz) waves from plasmas produced by intense femtosecond laser pulses has attracted much interest over several decades.<sup>1</sup> THz generation schemes that use a single-color single-pulse beam to produce a plasma are suitable for many applications because they are relatively simple to implement. The main target materials that have been proposed and studied so far include solids (thin foil<sup>2</sup>) and gases (noble gas,<sup>1</sup> air<sup>3</sup>). In comparison with a gas target, a solid target can more effectively provide THz waves because of its higher laser absorption;<sup>1,4</sup> 0.7-mJ THz waves with an energy conversion efficiency of  $1 \times 10^{-3}$  have been reported.<sup>2</sup> However, the difficulty of replenishing a solid target and the generation of large amounts of debris make it difficult to generate THz waves repeatedly. To realize the benefits of both solid and gas targets, we have previously proposed using atomic clusters to produce plasma for generating THz radiation.<sup>5</sup> Like gases, atomic clusters are debris-free and replenishable targets. We have previously reported that argon clusters can

provide THz waves with energies approximately 600 times higher than those from argon gas because of increased laser absorption.<sup>5,6</sup> However, for atomic clusters to be a feasible source of high-energy THz waves, further improvement of the energy of THz waves from a cluster plasma is necessary.

The interaction between a laser pulse and clusters has been studied for decades.<sup>7-13</sup> Zweiback *et al.* conducted time-resolved studies of the dynamics of cluster explosions and identified an optimal laser pulse duration that maximized the laser absorption.<sup>9</sup> Enhanced ion energies and X-ray yields have also been achieved by optimizing the laser pulse duration.<sup>9-11</sup> This raises the possibility of increased THz energy, but the relationship between laser pulse duration and THz wave generation is yet to be studied.

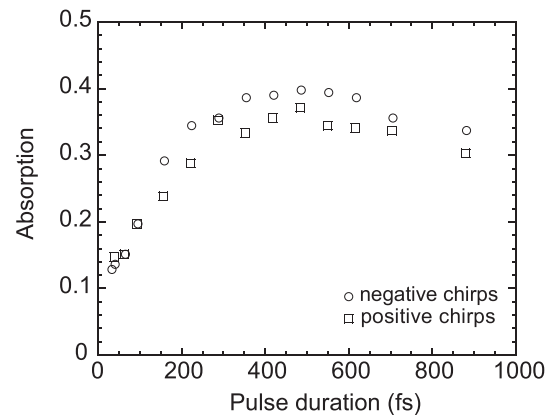
In this study, argon clusters were irradiated by a femtosecond laser with various pulse durations, and the energy of the resulting THz waves was increased by optimizing the laser

pulse duration. To understand the roles of the rising edge and the peak of a laser pulse, we irradiated argon clusters with a collinear double-pulse beam with various delay times. Using a double-pulse beam with the appropriate pulse interval also increased the energy of the THz waves.

The experiment was performed with a Ti:sapphire chirped pulse amplification laser system operating at a central wavelength of 810 nm. The laser pulse duration was controlled from 40 fs to 1 ps by changing the distance between a pair of gratings of the pulse compressor in the laser system. Pulses around the shortest pulse duration of 40 fs (full width at half-maximum) were positively or negatively chirped. The laser pulse was linearly polarized parallel to the optical table. The pulse energy was fixed at 3 mJ, and the laser intensity was varied according to the laser pulse duration. A plano-convex lens of focal length 200 mm focused the laser pulse onto the argon clusters in a Gaussian spot of diameter 10  $\mu\text{m}$  (full width at 1/e maximum). Argon clusters were generated by injecting 7-MPa argon gas into the center of a glass chamber made of fused silica glass with a transparency of 90% at 0.5 THz. The typical cluster radius was estimated to be  $\sim 5$  nm by Rayleigh scattering measurements.<sup>6</sup> The atomic density in the interaction region was estimated to be  $\sim 6 \times 10^{17} \text{ cm}^{-3}$ . Polystyrene foam and black polypropylene filters were inserted in the path of the THz waves to block undesirable residual laser pulse emission from the plasma. After a Tsurupica lens collimated the THz waves, their energy was detected by a helium-cooled InSb bolometer (QFI-2BI; QMC Instruments, Ltd., UK). The THz wave spectrum was measured with a Martin-Puplett interferometer.

In the double-pulse beam experiments, a beam splitter separated a laser pulse into two pulses, the first with an energy of 0.7 mJ and the second with an energy of 2.3 mJ. The polarization of each pulse was parallel to the optical table. The two pulses were focused onto the argon cluster collinearly with a delay time of 0–18 ps with a resolution of 0.03 fs. The THz detection system was the same as the one used in single-pulse beam experiments. THz energy was measured at an angle of 30° to the laser propagation direction.

We conducted an experiment to examine the reproducibility of the relationship between laser absorption and laser pulse duration. The absorption is defined as  $1 - I_t/I_0$ , where  $I_t$  and  $I_0$  are the laser pulse energies transmitted through the glass chamber with and without cluster generation, respectively. The laser pulse energy was detected with a pyroelectric energy meter on the laser propagation axis. Figure 1 shows laser absorption as a function of laser pulse duration. The laser absorption depends on the laser pulse duration and reaches a peak when the latter is  $\sim 350$ –550 fs. Thus, we show that laser absorption can be maximized by optimizing the laser pulse duration, which is consistent with previous findings.<sup>9,14</sup> This increased laser absorption is explained by resonant heating. For a spherical cluster, the resonant effect occurs when the electron density  $n_e$  is three times the critical density  $n_c$  (i.e.,  $n_e/n_c = 3$ ). If the plasma density just before the pulse peak reaches the cluster plasma satisfies this resonant condition, the resonant effect will occur. Therefore, the optimal pulse duration depends on when the

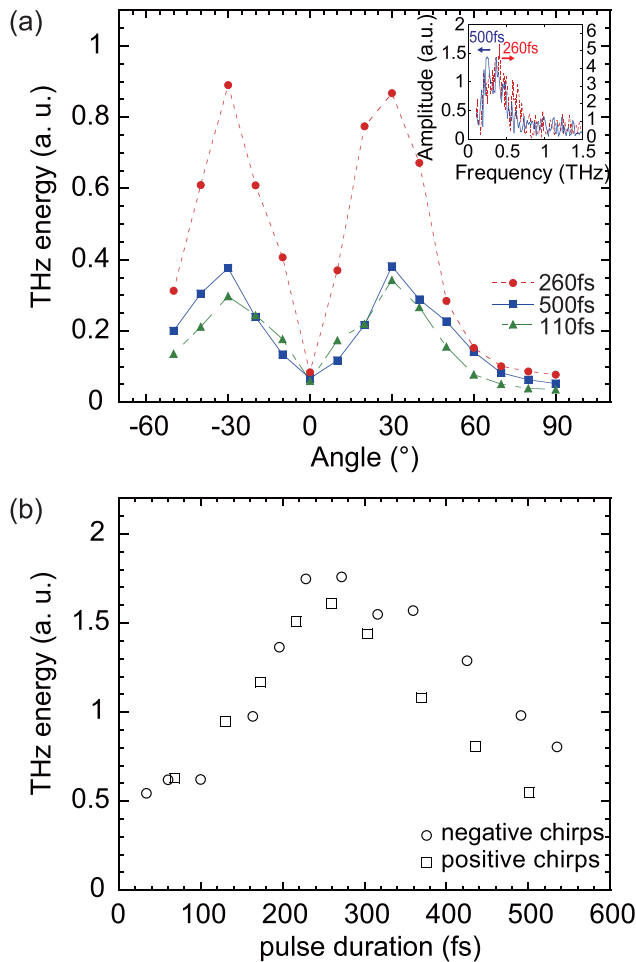


**FIG. 1.** Laser absorption of argon clusters for various laser pulse durations. Positive and negative pulse durations correspond to positive and negative chirp, respectively.

plasma density reaches the resonant condition.<sup>7</sup> We also show that the absorption for negatively chirped pulse irradiation is higher than that for positively chirped pulse irradiation; we discuss this later.

The dependence of THz wave generation on laser pulse duration was measured. Figure 2(a) shows the angular distribution of THz waves from the cluster plasma produced by a 3-mJ laser pulse with pulse durations of 110, 260, and 500 fs. The peak angles of the THz waves are around  $\pm 30^\circ$  to the laser propagation direction, irrespective of the laser pulse duration. The inset in Fig. 2(a) shows the spectral amplitude of the THz waves. The maximum frequency is  $\sim 0.8$  THz and the maximum emission is measured at 0.3–0.4 THz for the pulse durations of 110, 260, and 500 fs. However, compared with the spectral amplitudes for 110 and 500 fs, the one for 260 fs is slightly higher. Thus, we show that the energy of the emitted THz waves is increased by optimizing the pulse duration. The detailed relationship between THz energy and laser pulse duration at the angle of 30° is plotted in Fig. 2(b). The THz energy reaches its peak value when the pulse duration is  $\sim 250$  fs, which is somewhat lower than the pulse duration at which the laser absorption reaches its peak value (i.e., 350–550 fs). We attribute this difference to the effect of pulse duration on not only (i) plasma density, which is a key factor in resonant heating, but also (ii) laser intensity, which plays an important role in THz wave generation. Instead of changing the pulse duration, using double ultra-short laser pulses separates these two factors. Indeed, for a double-pulse beam, the pulse duration at which absorption is a maximum agrees with that at which THz wave generation is a maximum, which we discuss later.

Changing the laser pulse duration from 40 fs to 250 fs increases the THz energy roughly fourfold. Furthermore, a negatively chirped pulse generates THz radiation with higher energy than that produced by a positively chirped pulse, the difference being due to the rate at which the cluster plasma is heated. Assuming that a laser pulse imparts energy to the free

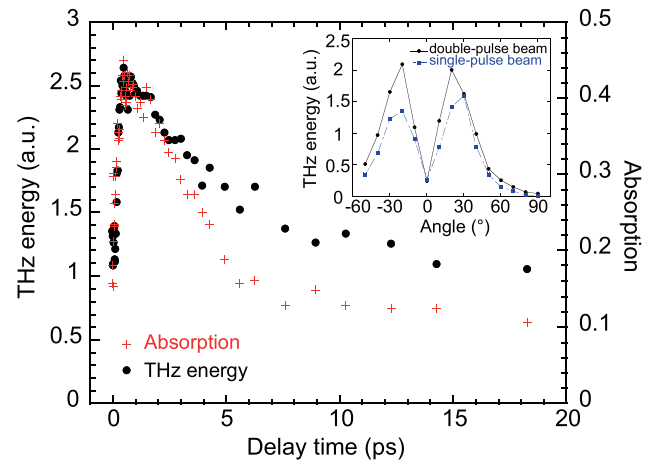


**FIG. 2.** Dependence of terahertz (THz) wave generation on pulse duration. (a) Angular distributions of THz waves for pulse durations of 110 fs (negative chirp), 260 fs (positive chirp), and 500 fs (positive chirp);  $0^\circ$  is the laser propagation direction. Inset: Spectral amplitude of THz emission for pulse durations of 260 fs and 500 fs with positive chirps. (b) THz energy for various laser pulse durations; the THz energy is detected at an angle of  $30^\circ$  to the laser propagation direction.

electrons in the cluster via collisional inverse bremsstrahlung, the heating rate  $\partial U/\partial t$  inside the cluster is

$$\frac{\partial U}{\partial t} = \frac{9\omega^2\omega_p^2\nu}{8\pi} \frac{1}{9\omega^2(\omega^2 + \nu^2) + \omega_p^2(\omega_p^2 - 6\omega^2)} |E_0|^2, \quad (1)$$

where  $\omega$  is the laser frequency,  $\omega_p$  is the plasma frequency,  $\nu$  is the electron-ion collision frequency, and  $E_0$  is the strength of the laser field in vacuum.<sup>7</sup> Fukuda *et al.* have demonstrated that, compared with a positively chirped pulse, a negatively chirped pulse heats a cluster more effectively and generates ions and electrons with higher energy.<sup>11</sup> This enhancement of the deposited energy increases the absorption and THz energy for negatively chirped pulse irradiation compared with positively chirped pulse irradiation.



**FIG. 3.** THz energy and laser absorption as functions of the delay time between the two pulses (pulse energies: 0.7 and 2.3 mJ); the THz energy is detected at an angle of  $30^\circ$  to the laser propagation direction. Inset: Angular distribution of THz waves for double-pulse beam (delay time: 0.87 ps) and single-pulse beam.

To demonstrate that optimizing plasma density can increase THz energy, we measured THz energy as a function of the delay time between the two pulses, the first acting as the rising edge of a pulse and the second acting as the pulse peak. The first pulse ionizes the clusters, and then the second pulse interacts with the cluster plasma,<sup>15</sup> the density of which is determined by the delay time between the two pulses. Figure 3 shows the laser absorption and THz energy for various delay times; the THz energy is detected at the angle of  $30^\circ$  to the laser propagation direction. Both the THz energy and the laser absorption are a maximum when the delay time is  $\sim 0.5$  ps. This clearly shows that the increase in the THz energy is related to laser absorption. Optimizing the delay time increases the number of electrons that generate THz wave as quadrupole radiation.<sup>6</sup> The difference between the optimal pulse duration and the optimal delay time is due to the different ways in which the atoms are heated. A pulse with the optimal pulse duration provides heating throughout its duration, thereby increasing the expansion speed and causing the resonant condition to be satisfied sooner.<sup>9</sup>

In summary, we have succeeded in increasing the energy of THz waves from a cluster plasma by optimizing the laser pulse duration. We attribute this increase to resonant heating that occurs when a pulse of optimal duration transfers energy to the cluster plasma. By optimizing the pulse duration, the THz energy and the absorption are both maximized. When clusters were irradiated with a collinear double-pulse beam, both the THz energy and the laser absorption were increased.

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